MEMOIRS

OF THE

COLLEGE OF SCIENCE

KYOTO IMPERIAL UNIVERSITY

SERIES A

VOL. XXIII, NO. 1

JANUARY 1940

KYOTO

1122-9

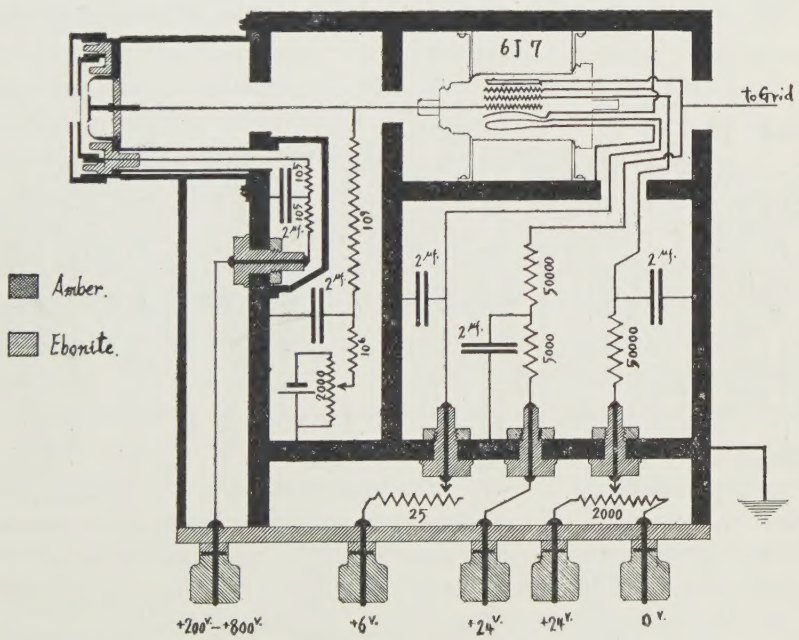
A Counting Instrument with Linear Amplifier

By Kiichi Kimura and Yoshiaki Uemura

(Received, October 28, 1939)

Recent development of nuclear physics has demanded the development of a new measuring instrument. A counter with linear amplifier was originally constructed by Greinacher[1] and then improved by Wynn-Williams and Wards[2]. Thereafter, several designs[3] were reported of amplifiers similar in principle to this one. In our laboratory too, the instrument was constructed with some improvements favourable to the study of nuclear transmutations[4]. Since, by careful construction and proper adjustment, the natural background of the count of this instrument is observed to be always less than one or two per hour in the case of the small ionization chamber described below, it is found to be most convenient for measuring the proton and alpha rays not only from strong sources but also from very weak ones, since even 3 of the particles per hour per cm^2 of the surface of an emitter may be ascertained.

Fig. 1



As the general description of an amplifier of this type is given by the writers cited above[1, 2, 3] it is only necessary here to describe the parts improved by the present workers.

a) Ionization Chamber

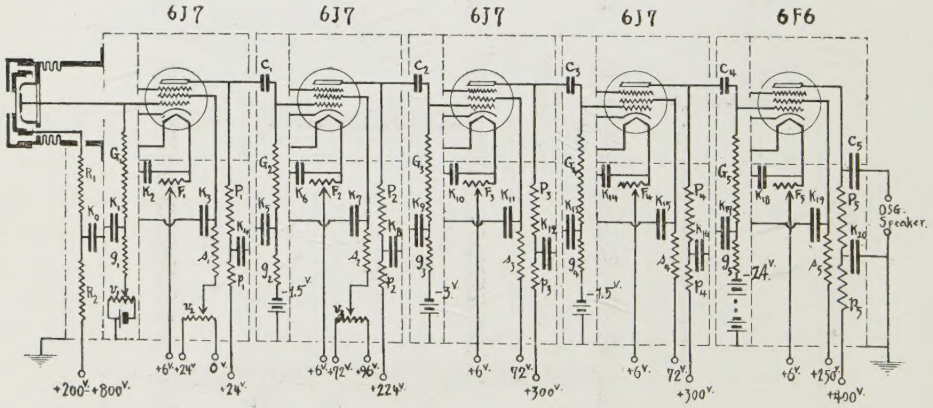
The counting chamber used (Fig. 1) is a parallel plate condenser having a collecting electrode 2 cm. in diameter. The inner electrode is surrounded at a distance of 1 mm. by a circular guard ring. The depth of the ionization chamber is made adjustable by screw between 10 mm. and 2 mm. (from the inner collecting electrode) and it has a circular opening 2 cm. in diameter covered by a thin sheet of aluminium or mica. The exterior of the outer electrode is shielded by an earthen cap with circular opening, which defines the effecting area of the source to be studied. This shielding is found to be indispensable for accurate measurement.

b) Linear Amplifier

The linear amplifier constructed by us is a form of resistance-capacity coupling. Having been used in several experiments[5] for long years, the function of this amplifier was successively improved so that it is used quite satisfactorily in the high voltage and crowded research laboratory. The circuit diagram is shown in Fig. 1 and 2 and the values of various components are indicated on the accompanying key.

i) *The first stage of the amplifier* As the most critical point of the

Fig. 2



$C_{1,2,\cdots,5}$	0.005 μF,	G_2	250,000Ω,	$P_{3,4}$	250,000Ω,
$K_{0,1,\cdots,20}$	2μF,	$G_{3,4,5}$	500,000Ω,	P_5	7,000Ω,
$R_{1,2}$	100,000Ω,	g_1	1,000,000Ω,	$P_{1,2,\cdots,4}$	5,000Ω,
$r_{1,2,3}$	2,000Ω,	$g_{2,3,4}$	20,000Ω,	P_5	2,000Ω,
$F_{1,2,\cdots,5}$	6Ω,	g_5	50,000Ω,	$s_{1,2,\cdots,4}$	50,000Ω,
G_1	10^8Ω,	$P_{1,2}$	50,000Ω,	s_4	2,000Ω.

amplifier comes in the first stage, the present workers, as have also all the others, have carefully laboured for this point. The valve of RCA-6J7 or UZ-77 (Tokyo Electric Company) was found to work satisfactorily (Fig. 1). This has, to a certain extent, desirable characteristics, i. e., low input capacitance, high amplification factor and non-microphonic construction. Because of its high noise level a valve of this type was considered by some workers unsuitable for the first valve, but this difficulty was overcome satisfactorily by proper control in the way described below.

After several tests it was found that the signal-to-noise ratio was approximately independent of the plate voltage over a wide operating range, but was closely dependent on the plate current as affected by control of screen grid voltage. The noise level could be greatly reduced by operating the valve at lower cathode temperature and with lower screen grid and plate voltage; for instance, a good condition was obtained by using a heating current of about 0.08 ampers, a plate potential of 24~48 volts and a screen potential moderately controlled between 12 and 24 volts.

In order to keep the amplifier always free from temporal impotence caused by some large electric disturbances and to gain high resolution of counting enormous numbers of particles in a short time without great sacrifice of sensibility, it is found desirable to use a grid leak resistance, at the first valve, of the order of 10^9 ohms with the grid voltage of about -0.8 volts which is the value of a floating grid. No remarkable increase of noise level was observed in this case as compared to the case of floating grid.

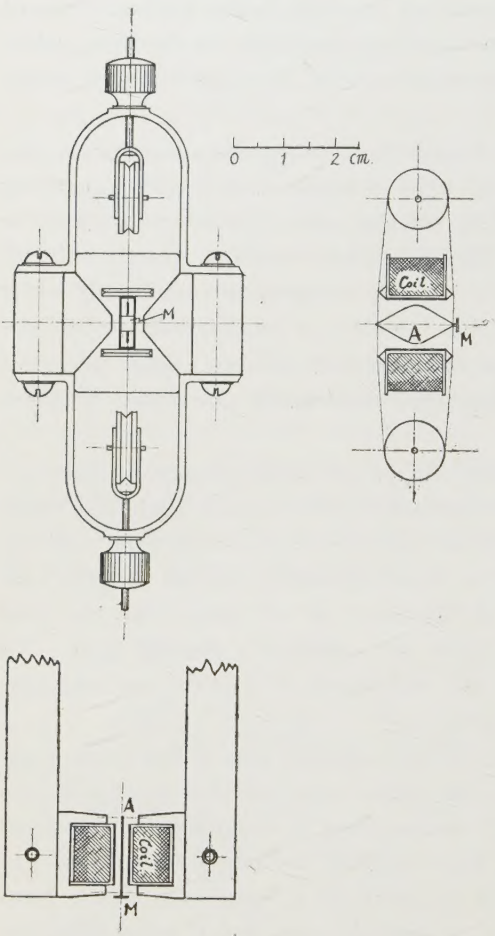
ii) *Shield case* The shield box of the amplifier was at first made from brass plate 3 mm. thick. But the brass plate of this thickness was found to resonate with sound waves from a neighbouring working machine and especially from a loud speaker connected parallel to the recording system itself; and so boxes of iron plate 1 cm. thick were substituted. The mere change of material was found very effective for obtaining good stability in the function of the amplifing system.

Each stage of the amplifier is mounted on a separate iron case which consists of three compartments; one for the valve, one for the filter circuit and one for the coupling circuit. To keep the valve from microphonic effect each valve has been suspended by soft springs with rubber sheet, and the connections were made entirely from flexible (dead beat) wires. In order to absorb mechanical vibration through the

table these iron boxes have been mounted on piles of wooden plates separated by pillars of sponge gum.

iii) *Control of amplification* As a slight change of the screen grid potential of the first valve has remarkable influence on the output of the amplifier, the control of amplification has been effected in this way without disturbing the lineation. This may be also possible at the second stage. It is desirable to control both complementarily, otherwise unwanted disturbances are experienced in the lineation. With other methods, published previously by several workers, this operation also may be a convenient method.

Fig. 3



c) Recording Apparatus

A simple oscillograph reconstructed from a balanced-armature-loud-speaker has been used. (This is preferable for observing the "kicks" since it records parallel to hearing.) A natural frequency of about 1000/sec may be obtained without difficulty. The construction was made following, in some respects, after Wynn-Williams[2]. (Fig. 3)

The soft iron armature A is a shallow rhomb in shape, the diagonals of which are 15 mm. and 8 mm. respectively. The vertices of the longer diagonal are suspended at the center of the speaker coil by two tungsten wires 80 μ in diameter forming sides of a loop which is stretched by means of a screw-and-pulley device. The distance between the two bridges of ebonite is 1.2 cm. For current coil, a speaker coil of 2000 ohms electrostatic resistance is satisfactory.

A thin galvanometer mirror M about 3 mm. square is attached to the armature. This whole part is immersed in liquid paraffin which acts as a damper. The optical system is mounted as usual.

Camera—A rotating paper (or film) camera was specially constructed so that a strip of highly sensitive bromide paper or photographic film stretched around a wheel is driven smoothly at various speeds by means of a phonomotor-and-pulley system. (Fig. 4)

The sensitivity of the oscillograph as usually operated was 1 mm. deflection per 2 volts at a distance of 60 cm. from the vibrator mirror.

d) The Characteristics of the Counting System

The lineation of the amplifying system and oscillograph as a whole was tested by inducing a known voltage of any frequency on the first valve grid and was found to be linear up to 1.5 cm. deflection at a distance of 60 cm. from the vibrator mirror. By using polonium source the Bragg's ionization relation was tested and found to be satisfactorily fulfilled.

The practical examples are reproduced in plate I.

In conclusion the authors wish to express their cordial thanks to Prof. Dr. B. Arakatsu for his kind guidance and valuable suggestions throughout this work, and at the same time their thanks are due to Hattori Hokokwai and Nippon Gakujutsu Sinkokwai for financial support in this work.

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Explanations of Plate I.

a——α-particles from the natural surface of Hokutolite.
The thickness of absorber: 13 mm. in air equivalent.
The depth of the ionization chamber: 7 mm.

b——α-particles of Po deposited on a copper plate.
The thickness of absorber: 20 mm. in air equivalent.

The depth of the ionization chamber: 2 mm.

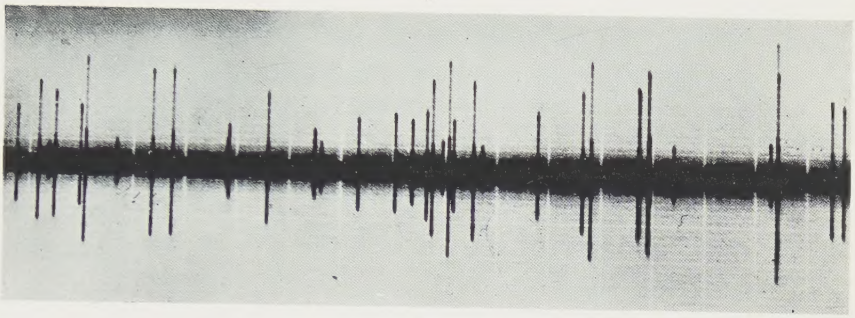
c——α-particles of Po deposited on a copper plate.
The thickness of absorber: 30 mm in air equivalent.
The depth of the ionization chamber: 2 mm.

d——Homogeneous α-particles (12.7 cm) by the reaction: $Li^6+D^2\rightarrow 2\,He^4$. (thick Li_2O-target).
The thickness of absorber: 8.3 cm. in air equivalent.
The depth of the ionization chamber: 2 mm.

e——Homogeneous α-particles (4.5 cm.) by the reaction: $B^{11}+H^1\rightarrow Be^8+He^4$ (thick borax target).
The thickness of absorber: 4 cm. in air equivalent.
The depth of the ionization chamber: 4mm.

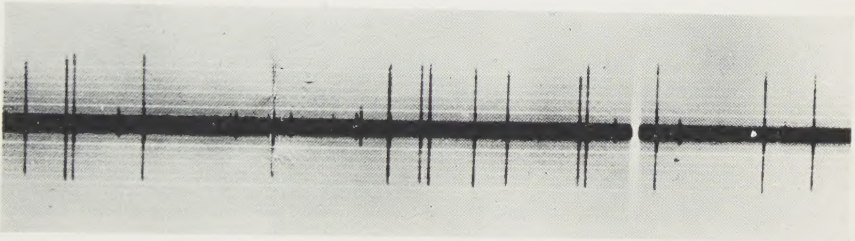
Plate I

a

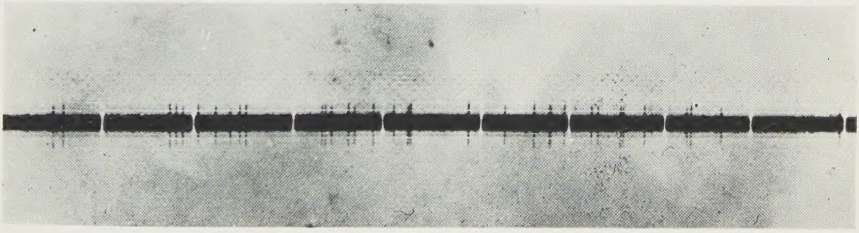


b

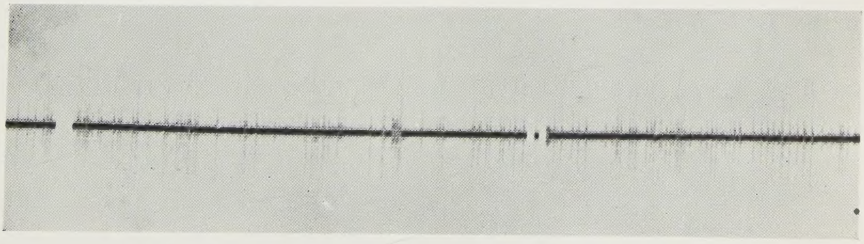
c



d



e



Study on Radioactivity of Hokutolite in Taiwan by Means of a Counter with Linear Amplifier

By Kiichi Kimura

(Received, October 28, 1939)

I. Introduction

For the purpose of research in radioactivity of minerals, the intensity has generally hitherto been measured by using an electrometer, Geiger point counter or Geiger-Müller cylindrical counter. These all demand complicated procedures lasting for a number of days to decide the kind of rays (i. e., α-, β- or γ-rays) and to measure the contents of radioactive element in any sample. Practically, as they also suffer cosmic and local γ-ray effects, it is not easy to detect minute quantities of radioactive substance unless a chemical process is combined with them. These complexities are well circumvented by using a counting system with linear amplifier[1], which makes it possible to detect individual α-particles in the presence of β-and γ-rays. Thus accurate measurements may be carried out for such feeble α-emitter as igneous rocks. The author confirmed the superior efficiency of this counter in his study on α-radioactivity of Hokutolite and obtained a number of interesting results. Especially, the observation of the activity from part to part of a rock furnished us new knowledge concerning the essential properties of this mineral.

II. Historical

The radioactivity of Hokutolite was first discovered by Okamoto and Sunohara[2] on the river bed, within a limited range, of Hokuto-kei in Taiwan (Plate I). This is a spring-sediment which is deposited at most up to 2 cm. in thickness on the surface of andesite or sand-stone and is composed of an aggregate of granular rhombic crystals of white or light gray colour (Plate II a).

Among the results obtained by Okamoto and Sunohara[3] by means of prolonged photographic action, the following properties of the activity are of particular interest. i) The activity of the surface of any specimen is far stronger than that of its interior. ii) Regarding the surface activity, that which has distinct crystals is stronger than the

rest, and that of white colour is stronger than that of gray. iii) The activity of any surface does not decay during a period of several years.

Later, Hayakawa and Nakano[3] reported that they chemically separated ionium, radium and polonium from the sediment and observed the alpha-activity by a fontactoscope.

Then they considered the mechanism of the sedimentation of the mineral and concluded that the ionium only is the mother element from which all other radioactive materials are produced.

The results described in the present paper are derived by a new method different from that of preceding workers on the problem and are considered to be more reliable in some respects than their results.

III. Collections of Hokutolite

By permission of Taihoku Province collections of Hokutolite in Hokuto-kei were made several times between October 1935 and December 1938.

The main object of these collections at first was to provide a source of homogeneous alpha-rays, such as polonium if possible, which might be used for the standardization of a counting instrument, for we had none of the source for use at that time. For this purpose some twenty different sediments were collected, and their alpha-activities were detected by the counter with linear amplifier[1].

By this counting method it was easily found that the sample which was an aggregate of white colour crystals was most intense and one which was of distinct crystals was more intense than other samples. This result agrees with that of observations made earlier by Okamoto. These two kinds of aggregates of crystals were found, in general, dried on the dry stream-bed or in stagnant water ; while others, such as some with a smooth surface, were under the flow of the spring. With this knowledge a great amount of specimens which had intense activity was collected, and polonium source of sufficient intensity was chemically prepared from them.

IV. The α-Activity of Natural Surface and that of Interior of Minerals

By preliminary experiments it was easily found that while the intensity of surface activity of various specimens showed distinct difference over a wide range, the activity of the interior of all of the samples was observed to be of almost equal intensity.

By bringing the sample up to 3 mm. before the window (1 cm. in diameter) of the ionization chamber[1] (5 mm. in depth), the aperture of which was covered by Al foil of 1 cm. equivalent of air, the number of α-particles emitted from the natural surfaces of various samples was counted. It was found to be 180/min. to 50/min., while that from interior of them was in all cases about 13/min. in contrast to the wide difference found in the activity on their natural surfaces. The same conditions were found in a sample* which is supposed to have been collected about 30 years ago. (This does not mean that the last sedimentation took place 30 years ago). The above results are summarised in Table I.

Table I.

Relative number of α-particles/Min.							
Sample No.	1	2	3	4	5	6	
Surface	180	130	128	74	50	52	mean 13.5/min.
Interior	14	13.2	13 3	14	13.5	13	

Table I. No. 6 is the old sample in the Museum of Taiwan. No. 2, 4 and 5 are for smooth surfaces: others are for zig-zag surfaces.

From these results we may conclude that in the interior the activity for all different specimens is constant within statistical fluctuations. Thus the distribution of radio-elements seems to be uniform in the interior of all specimens, and certain radio-elements of long half period are probably in equilibrium with their products, while in the surface there may be some element of relatively short life deposited. Accordingly it seems wise to interpret the variety of intensity of surface activity as due to the age of the specimen rather than to its irregular contents. The justification of this interpretation will be made in a later chapter after the nature of the radio-elements included in the sediment has been studied.

V. Intensity Variation with Depth from Natural Surface

If an intensity variation of the activity corresponding to different depths under the surface is known, the above results will be further confirmed and then the speed of sedimentation can be roughly estimated.

* This had been exhibited in the Museum of the Governor-General of Taiwan, by whose kind loan these results were verified. The author here expresses his thanks to the staffs of the Museum.

For this purpose a sample, No. 4 in Table I, (collected in a fast stream) was chosen, as this surface was fortunately almost plane and smooth. The α-particles were counted for each successive new surface after it had been carefully cleaned with a hard brush in running water so as to remove any powder of the preceding surface. The results are shown in Table II and Fig. 1.

Fig. 1

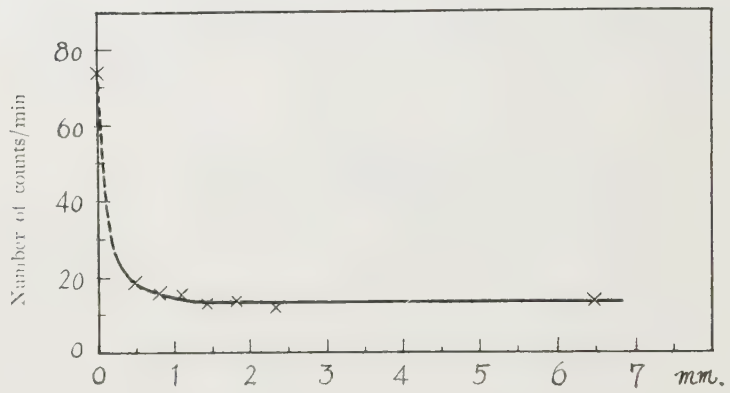


Table II.

Depth under the surface in mm.	0	0.5	0.8	1.1	1.43	1.83	2.33	6.5
Number of Counts/min.	74	19	16	16	13	14	12	14

Table II. The diameter of the window of the ionization chamber is 1 cm.

In this sample, the equilibrium comes at about 1.3 mm under the surface and then continues for further depth within the statistical fluctuations. As this result corresponds fairly well with that of Table I, it may be admitted to be true in general. Almost the same aspect has been obtained too, by the photographic action of α-particles (Plate II b). The black points which appear like sun-spots in the Plate II b are due to weak activities of the points which were probably worn down by some cause. It may be therefore conceivable that the growth of Hokutolite has been going on either exceedingly slowly, or intermittently in the course of a long time at the period when favourable conditions prevailed. But the latter explanation is more probable since

the temperature and chemical components of the spring suffer irregular variations from the meteorological conditions.

VI. Determination of the Kind of Radio-elements

Now, if we want a satisfactory explanation of the interesting aspects of radioactivity, it is necessary to make clear the kind of radio-elements

Table III.

Natural Surface

Aluminium in mm. air range	Number of counts per 10 min.	Aluminium in mm. air range	Number of counts per 10 min.
10	2310	45	201
15	1962	50	156
20	1603	52	126
25	1217	55	110
30	863	60	75
32	710	62	57
35	504	65	19
40	372	70	16
42	282	72	14

Table III. The diameter of the window is 1.5 cm. The distance between the source and the window is 2 mm. The depth of the chamber is 10 mm.

Fig. 2

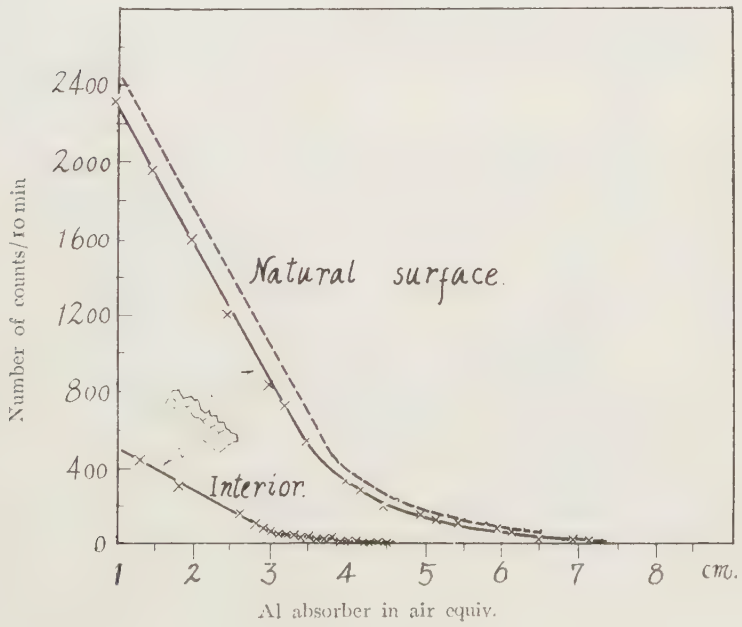
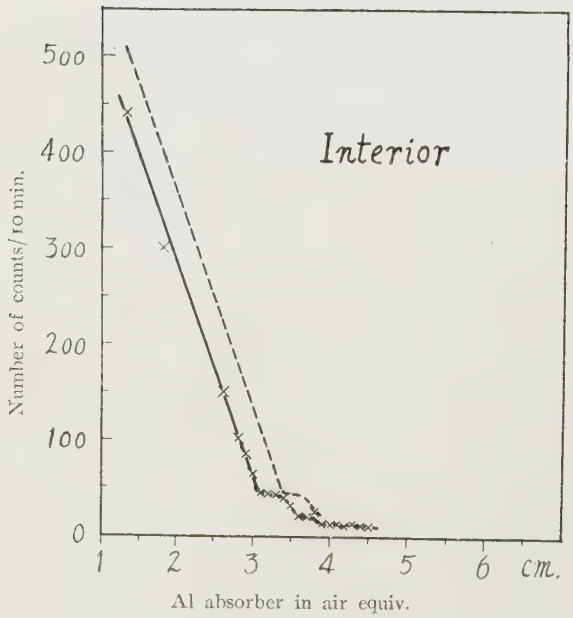


Table IV.

Interior			
Aluminium in mm. air range	Number of counts per 10 min.	Aluminium in mm. air range	Number of counts per 10 min.
13	438	35	35
18	308	36	20
26	150	37	24
28	106	38	32
29	86	39	14
30	66	40	16
31	53	41	14
32	46	42	10
33	46	43	14
34	40	44	8
		45	8

Table IV. Experimental conditions are quite the same as in Table III.

Fig. 3



contained in the sample.

For this purpose the number of α-particles from the natural surface of a sample and also from the interior was measured for various thicknesses of aluminium foil inserted between the ionization chamber and the sample.

The results are shown in Table III and Fig. 2, and Table IV and Fig. 3 for surface and interior respectively. In this case the diameter of window was 1.5 cm., the distance between source and window was 2 mm. (earthen cap was excluded), and the depth of the ionization chamber was 10 mm. For the purpose of comparing the activity of the surface to that of the interior, the number of counts at

the interior is given annexed to that of the surface in Fig. 2. We see a) that the activity of the surface is far stronger than that of the interior and b) that the range of α-particles i. e. the kind of radio-elements is quite different in the two cases.

As the abscissa of Fig. 2 (and 3) gives only the thickness (in air range) of Al foil inserted between the source and the window of the detector, the true curve should be displaced some 2 mm. (air part) to the right as shown in the broken curve. The end of each straight curve represents the range of α-particles, i. e. *ca.* 3.8 cm. and *ca.* 3.3 cm. in Fig. 2 and Fig. 3 respectively. The former is very near to the range of Po-α-particle and the latter to that of Ra-α-particle.

It is natural to conclude that the activity of the young natural surface is due to mainly polonium and that of the interior to radium and its products. This explanation accounts for the various facts we have mentioned concerning the activity of Hokutolite. Moreover, by the curve in Fig. 2 the presence of some long range particles in small amount is recognized. These are probably some members of the thorium series, although Hayakawa and Nakano[3] were convinced that none of these were present.

This method is certainly not a refined one for analysing radio-elements, but it is a rather convenient one for our purpose.

The ordinary accurate method is to measure the range-number curve by using the source of fine powder deposited on a glass plate by floating method, but experiment in this line has not yet been carried to a conclusion.

VII. Estimation of the Contents of Both Radium and Polonium

It is now possible to make a rough estimate of the radium and polonium content, though there can be no accurate measure of the polonium content because its value diminishes as a function of time.

Thin sources of fine powder granulated from the surface and the interior respectively of a sample, the activity of which was relatively strong, were prepared on glass plates by the floating method with ethyl alcohol. The thickness of each was 5.5 mg/cm^2. Bringing this source up to 3 mm. from the window of the ionization chamber, the depth of which was 10 mm., we counted the number of particles by insertion of an aluminium sheet of 1 cm. air equivalent. In this case the activity of U_3O_8 was used as standard. The numbers of counts per 1 mg per 10 minutes are as follows;—

$(U_{I,II})_3O_8 \sim 847$, hence for 1 mg $U_{I,II}$
$U_{I,II} \sim 1016$/mg/10 min. (1),
Simultaneously for Hokutolite { interior ~ 8 (2), surface ~ 35.4 (3).

By Geiger and Lord Rutherford[4], the total number of α-particles emitted by $U_{I,II}$ was found to be 2.37×10^4/g/sec. i.e. 14220/mg/10 min. The present values should also be multiplied by about 14 so as to estimate roughly the total number of α-particles emitted,

that is, for Hokutolite { interior ~ 112/mg/10 min. (4), surface ~ 496/mg/10 min. (5).

As in the interior the α-activities are mainly due to Ra and its four products, at least, up to RaC′ in equilibrium (see VI), 28 in 112 are due to Ra. Then we have radium contents of 1.3×10^{-9} g per 1 g of Hokutolite, by using 3.72×10^{10} particles per 1 g of Ra per second. On the surface $496 - 112 = 384$ particles per mg per 10 min. are mainly due to Po and the upper limit of concentration of this is 3.8×10^{-12} g per 1 g of the surface material, as 1 g of Po emits 10×10^{17} particles per 10 minutes.

Since 1.3×10^{-9} g Ra equilibrate with 2.6×10^{-13} g Po, the content of Po on the surface of Hokutolite is more than 15 times that of the direct derivatives of Ra.

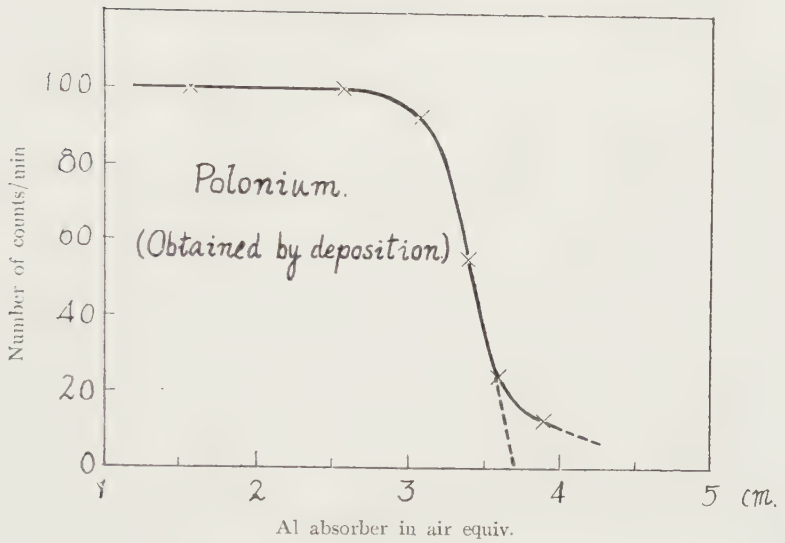
This result is very significant and shows that the activity of Hokutolite comes from two mother elements Ra and probably RaD which are thought to be deposited as sulphates in crystals of the mineral with the elements Ba and Pb.

VIII. Chemical Separation of Polonium

As the content of Po on the natural surface seemed to be large enough to provide the source of homogeneous α-particles, chemical separation was performed as follows:

After boiling about 100 g of fine powders of mineral with sodium-carbonate, the melt was thoroughly washed with water and dissolved in dilute hydrochloric acid. Immersing copper (or nickel or silver in some cases) plate in this solution for about 20 hours, we found Po—a strong α-active substance—deposited on the plate. The number of counts was 1958/min. By carefully adjusting the counting system, it was found that the α-particles emitted by the deposit are homogeneous as shown in Plate III a, in which the deflections are uniform, while those by the α-particles from natural surface are irregular (Plate III b). The range curve is given in Fig. 4.

Fig. 4



IX. Discussion (Process of Sedimentation)

From the above experimental results we see that the alpha-activity of Hokutolite is mainly due to polonium and radium with radium products. The radioactive equilibrium from radium up to radium C′ takes place after about 40 days starting from pure radium. Further most of the polonium, especially on the natural surface, is not a direct derivative of radium, because in respect to intensity the former is far stronger than would be expected from the equilibrium relation with the latter. Since an equilibrium from radium to radium D and consequently to polonium comes after about 110 years,* it is difficult to consider such intense activity of polonium on the young surface as due to radium. If we now take radium D which has a half period of 16 years* and equilibrate after 800 days* with polonium as mother element, the activity of the latter should then reach maximum and slowly decay keeping equilibrium with radium D. This character can not be proved by taking up real examples one by one, but may be theoretically argued from the following facts: (1) The activity on the surface does not seem to decay markedly for a number of years and (2) that of an old sample probably collected 30 years ago has diminished to less than half

* St. Meyer and Schweidler, Radioaktivität, p. 456.

that of a young one. Therefore samples of opaque and white colour, which show stronger activity than those tnat are gray and somewhat transparent (which is taken to be young), may probably be in a state of maximum intensity being in equilibrium with radium D. But on this point more samples and a longer period of observation are necessary to give complete explanation.

Now with this knowledge and the results of chemical analysis of both specimens and the spring water we can picture to ourselves the process of sedimentation of Hokutolite.

By chemical analysis the spring water was found to be strongly acid and to contain much sulphate with a very small amount of lead and barium. As Ishitani[5] pointed out, the spring water contains a large amount of radium emanation and so it may be inferred that radium is deposited with barium as sulphate on the surface of mother stone (i. e. andesite or sand stone), and radium D simultaneously with lead as sulphate, since all the daughter elements of radon soon change to radium D for their short half periods.

Other series, presumably a faint amount of thorium series, may possibly be contained in the surface as Fig. 2 suggests; but if so, since the half life of surface activity is short, they must be mesothorium (half period is about 6 years) and its products.

X. Summary

In the present work the author observed the radio-activity of α-particles of Hokutolite by a counter with linear amplifier and obtained the following interesting results:

1) The activity of the natural surface of young mineral is far stronger than that of the interior; for example, we have a sample in which the former is more than 10 times the latter.

2) The wide variation of the activity of surfaces in various samples, in spite of their being almost constant in the interior, is due not to difference in constituents of the specimens of Hokutolite, but to the lapse of time after the last sedimentation took place.

3) α-activity of the surface is mainly due to polonium (3.8×10^{-12} g) and that of the inside to radium (1.3×10^{-9} g) with its products being in equilibrium. The content of Po is more than 15 times that of direct derivatives of Ra.

4) Considering (2), it seems a large time-lag exists between the period of sedimentation of surface and that of interior at most 1 mm.

beneath it. Consequently, the sedimentation of this mineral must have been going on very slowly, or intermittently when favourable conditions prevailed.

5) Finally, the process of sedimentation has been found to be as follows ;—

i) The direct mother element of polonium in Hokutolite is RaD- the derivative of radon considerably contained in the spring.

ii) While $RaSO_4$ in the spring is deposited withB aSO_4, RaD SO_4 or $RaBSO_4$, directly derived from radon, crystalises out with $PbSO_4$.

Acknowledgement

In conclusion the writer wishes to express his sincere thanks to Prof. Dr. B. Arakatsu for his many helpful suggestions and valuable advice so generously given throughout the course of his work ; and to Prof. Dr. I. Hayasaka of Taihoku Imperial University for his kind support in collecting samples of Hokutolite. He is also indebted to Mr. M. Naito and Mr. Y. Uemura for their help in this work. Lastly his thanks are due to Hattori Hokokwai for financial support which rendered the study possible.

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5. Ishitani, Toyo-Gakugei-Zassi, **367**, 159 (1912).

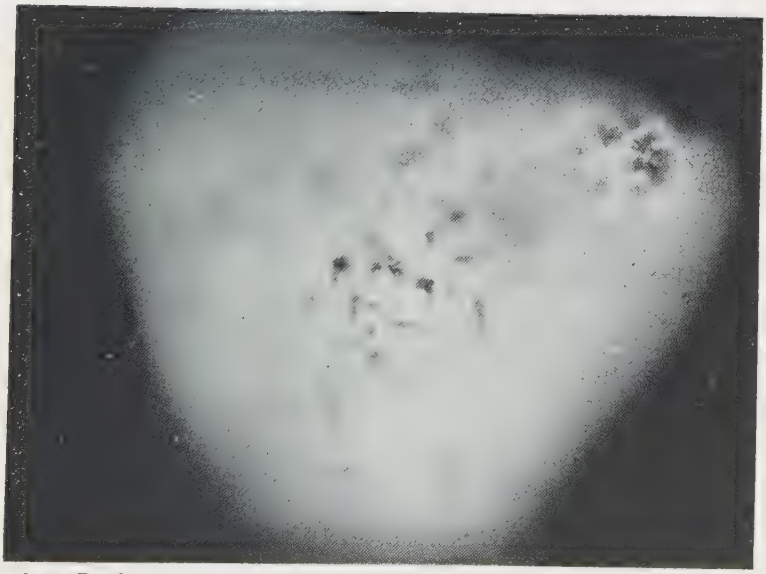
Plate I

a ——A part of Hokuto-kei

b——A large stone covered by Hokutolite of strong activity.

Plate II

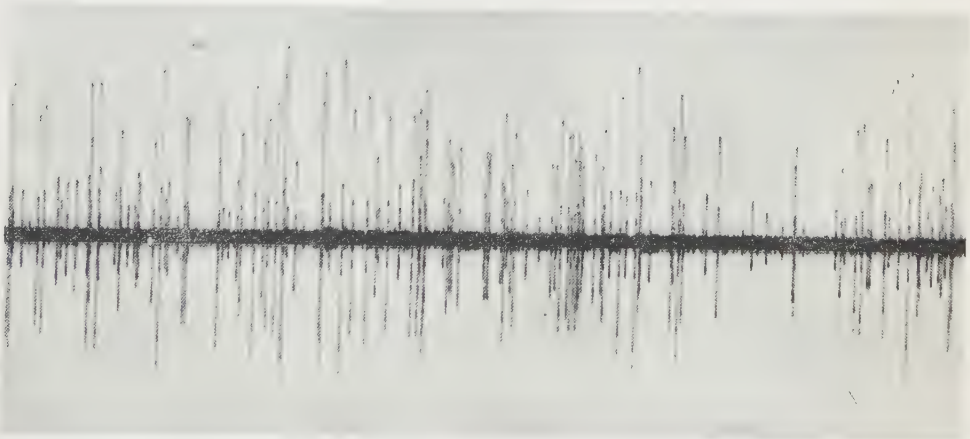
a——Hokutolite



b——Blackening of photographic plate by Hokutolite through Al sheet of 1/100 mm. Exposure is 50 days.

Plate III

a——Polonium



b——Hokutolite

Liberation of Fast Neutrons in the Nuclear Explosion of Uranium Irradiated by Thermal Neutrons*

By Tokutaro Hagiwara

(Received October 31, 1939)

Abstract

Fission neutrons liberated from uranium induced by neutron irradiation were studied concerning the *separated* effect due to thermal neutrons alone. Uranium oxide was either screened from primary thermal neutrons or exposed to them by use of a cadmium sheet shield interposed between the neutron source and uranium oxide. The comparison between the photographically recorded counts of neutrons per definite time interval measured with and without the cadmium shield allowed us to detect and measure the net gain in the number of the fast neutron release from uranium. By a number of sets of such alternating measurements it was possible to ascertain the remarkable *net increase* of the secondary fast neutrons caused by absorbing the thermal neutrons alone. In this way, the average number of the fast neutrons liberated per fission of uranium induced by thermal neutron was found to be *2.6*. From the considerations of the order of the amount of energy—about 10 Mev.—involved in the fission neutrons, it is probable that the neutron release takes place *immediately* after the main division of the compound nucleus instead of simultaneously with the explosion.

As was first discovered by Hahn and Strassmann,[1] uranium under neutron irradiation is split by absorbing neutrons into two lighter elements of roughly equal weight and charge, the division being accompanied by an enormous amount of energy release. It follows, then, that these fission fragments produced would contain *considerable excess* of neutrons**as compared with the corresponding heaviest stable isotopes with the same nuclear charges, provided that the division takes place only into two parts. In fact, a part of these excess neutrons was

* A short description of this work has been published in the *Review of Physical Chemistry of Japan*, **13**, 145 (1939).

** For example, the assumption of a division of $^{239}_{92}U$ into $_{56}Ba$ and $_{36}Kr$ leads to the expectation of an excess of about 15 neutrons. The excess of these neutrons should be got rid of by possible processes to bring back to the neutron-proton ratio of the stable nuclei.

1. O. Hahn and F. Strassmann, Naturwiss., **27**, 11 (1939).

2. Numerous β-*active* products, for example, with atomic numbers lying between *35*(*Br*) and *57*(*La*) were revealed by chemical work.

I. Curie and P. Savitch, Compt. rend., **208**, 343 (1939). O. Hahn and F. Strassmann, Naturwiss., **27**, 89 and 163 (1939). P. A. Heyn, A. H. W. Aten jun. and C. J. Bakker, Nature, **143**, 516 (1939). P. Abelson, Phys. Rev., **56**, 1 (1939) and numerous subsequent reports.

found to be disposed of by the subsequent β-ray transformations of the fission products,[2] but another reasonable possibility of reducing the neutron excess seems to be associated with a *direct emission* of the neutrons, which would either be emitted as a part of fission products at the moment of the nuclear splitting or escape from highly excited nuclei of the fission heavy fragments after the main division of the compound nucleus. It may, therefore, be supposed that the fission process results in the liberation of more than one secondary neutron per nuclear splitting.

Following the discovery of this new type of reaction, it has been attempted by several workers[3–8] to observe this pronounced emission of neutrons from uranium irradiated by neutrons. It is now of interest to find more accurately whether and to what extent the emission of neutrons takes place in uranium by absorbing neutrons, especially *thermal* ones,[9] and also to estimate the number of the neutrons released per fission in connection with the possibility of a cumulative process of exo-energetic transmutation chains.

In some of the previous experiments[3,4,5,8] a comparison was drawn between the numbers of thermal neutrons present in a large water tank with and without uranium under the irradiation of photo-neutrons from a Ra γ -Be source at the centre of the tank, and this comparison was made by measuring the activity induced in neutron detector in the water. On such a principle, von Halban, Joliot and Kowarski[3] have been able to observe an increase of about five percent for the thermal neutron density in the water, when uranium was present in its aqueous solution, and they have estimated the corresponding yield to be 3.5 neutrons per fission. Anderson, Fermi and Szilard[5] have also quite recently reported a striking increase of even about ten percent, in the case when about 200 kg of U_3O_8 was arranged around the photo-neutron source of 2.3 grams Ra situated at the centre of a vessel containing a large amount of ten percent aqueous $MnSO_4$ solution, the activity induced in manganese being taken as a measure of the number of thermal neutrons present.

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3. H. von Halban, F. Joliot and L. Kowarski, Nature, **143**, 470 and 680 (1939).
4. H. L. Anderson, E. Fermi and H. B. Hanstein, Phys. Rev., **55**, 797 (1939).
5. H. L. Anderson, E. Fermi and L. Szilard, Phys. Rev., **56**, 284 (1939).
6. L. Szilard and W. H. Zinn, Phys. Rev., **55**. 799 (1939).
7. G. von Droste and Reddemann, Naturwiss., **27**, 371 (1939).
8. J. L. Michiels, G. Parry and G. P. Thomson, Nature, **143**, 760 (1939).
9. It has been suggested by Bohr—*Phys. Rev.*, **55**, 418 (1939)—that the fission process caused by the *thermal* neutrons is to be ascribed to the neutron capture by the rarer isotope ^{235}U rather than the abundant one ^{238}U.

Since in those experiments (made in such a system as a mixture of uranium and water) part of the neutrons are considered to be absorbed into uranium and to induce the fission process before reaching down to thermal energies, it seems desirable to make measurements on the *separated* effect due to thermal neutrons alone. The object of the present experiment is to obtain some information regarding a net increase in the number of fast neutrons released from uranium by absorbing the primary thermal neutrons alone.

Experimental Method

In order to ascertain a net gain in the number of secondary neutrons from uranium caused by the action of thermal neutrons, we carried out the following experiments. The experimental arrangement is shown in Fig. I.

To obtain thermal neutrons, a cylindrical paraffin block, 20 cm in diameter and 25 cm in height, was made, in the central cavity of which was placed the usual neutron source of *50 mg radium α-beryllium* enclosed in a platinum container.

The container of uranium oxide powder was a double walled, cylindrical copper vessel, 25 cm in the outer diameter, 21 cm in the inner diameter and 25 cm in height, and the cylindrically zonal space between the double walls contained the fine powder of U_3O_8. The paraffin block (with neutron source) was placed inside the innerwall of this vessel.

The outer surface of the vessel was completely covered on all sides by the cadmium sheet shields 1 mm thick, which prevented thermal neutrons from escaping to affect the detector. An *inner cadmium* sheet cylinder (about 20.5 cm in diameter) was also placed, if desired, so as to cover the whole cylindrical surface of the paraffin block, and served as a shield to prevent the slowed down source neutrons from interacting on uranium. Accordingly, uranium oxide could be either *screened* from the action of the thermal neutrons or *exposed* to it by simply setting this inner shield in place or taking it away.

As a detector for slow neutrons, a *boron* cylindrical ionization chamber, 2.2 cm in diameter and 20 cm in length, lined with a thin layer of boron was used. The ionization pulses from the chamber were fed to a grid of a linear amplifier of Wynn-Williams-type,[10] through

10. B. Arakatsu, K. Kimura and Y. Uemura, Memoirs of the Faculty of Science and Agriculture, Taihoku Imperial University, **18**, 83 (1936).

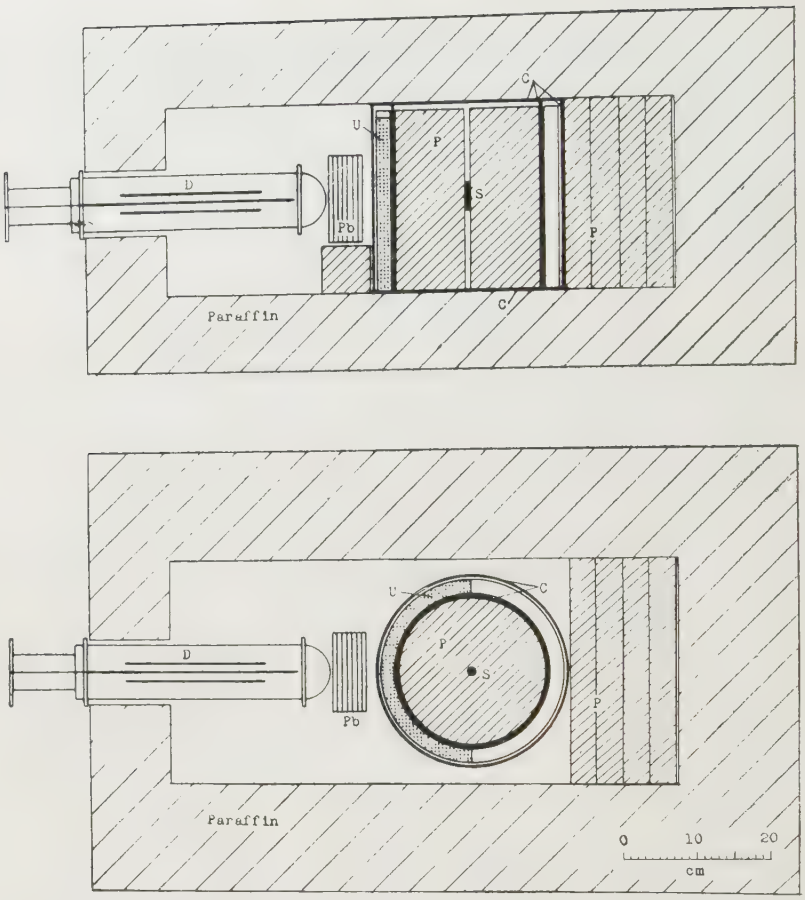

Fig. I. Vertical and horizontal sections through centre of the arrangement. S, Neutron source. P, Paraffin wax. C, Cadmium sheet shields. U, Uranium part filled with uranium oxide. Pb, Lead block shield. D, Cylindrical ionization chamber.

which the frequency of these pulses was counted from the records of kicks on the moving sensitized photographic papers by means of an ordinary oscillograph. In order to reduce the background effect due to γ-rays from the source, a disk of lead shield (5 cm thick in the axial line of the ionization chamber) and 12 cm in diameter was interposed between the uranium container and the head of the horizontally placed ionization chamber. The whole apparatus was placed in a thick walled (about 15 cm) paraffin chamber, the inside dimensions of which were $30 \times 70 \times 26$ cm, as shown in the accompanying diagram.

The present method of neutron detection is eventually based on measuring the number of the neutrons in question as that of only the neutrons which have been slowed down on the whole inner walls of the enclosing paraffin chamber after passing through the outer cadmium shields with relatively high velocities. So a simple comparison of the counts of neutrons per definite time interval measured with and without the inner cadmium shield, the outer cadmium shield being in place, would allow us to detect and measure the net increase in the number of relatively fast neutrons inside the outer cadmium cylinder. If any positive increase exists, it must be ascribed to the secondary emission of (relatively fast) neutrons from uranium by the effect (presumably fission effect) of thermal neutrons, because no appreciable effect of the presence or absence of the inner shield could be detected, when the vessel was empty and the outer shield (1 mm thick) was placed in its position.

Liberation of Secondary Neutrons from Uranium

A number of such *alternating* measurements (in the presence and absence of the inner cadmium shield) were made per 10 minutes interval on the oscillograph records, when one of the two vertical *semi-circular* halves, into which the space between the double walls of the cylindrical vessel was partitioned, was filled with *2.5 kg* of uranium

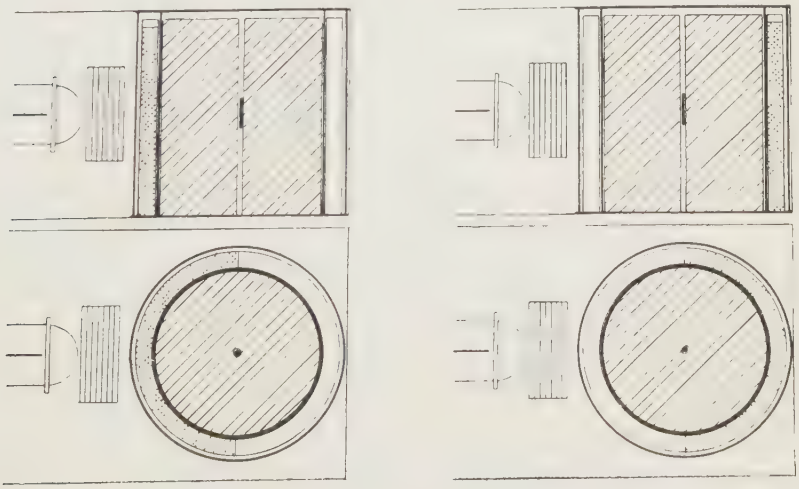

Fig. II.

Arrangement i

Uranium part is placed close to the ionization chamber.

Arrangement ii

Uranium part comes on the side opposite to the ionization chamber with respect to the neutron source.

oxide. The semi-circular uranium part was placed close to the ionization chamber as shown in Fig. II.-*i*. The front of the outer wall of the uranium part was kept, across the lead shield block, at a distance of 6.4 cm from the head of the cylindrical ionization chamber in its axial direction.

The number of recorded counts per 30 minutes was *C-i* (=*2166*, in Table I), when uranium oxide was exposed to the thermal neutrons (in the absence of the inner cadmium shield); while it was reduced to *D-i* (=*2117*, in Table I), when uranium oxide was screened from the thermal neutrons with the inner cadmium shield. The difference, *49*, between these two numbers must be ascribed to the fast neutrons emitted by uranium under the irradiation of the thermal neutrons alone.

Under such a geometry (i) of the arrangement as described above, however, only a fairly slight increase of neutrons was observed, and so the measurements were tried again under another geometry (ii) of the arrangement. 2.5 kg of uranium oxide was now put into another semi-circular zonal part of the vessel as shown in Fig. II.-*ii*., so that the uranium part came on the side opposite to the ionization chamber with respect to the neutron source and it was, at its remotest part, farther away by 25 cm (the outer diameter of the uranium vessel) from the ionization chamber, namely, the front of the part was withdrawn to 29.4 cm from the head of the ionization chamber in the axial line.

Under such a geometrical condition (ii), measurements similar to the previous ones were carried out. A count *C-ii* (=*2261*, in Table

Table I

	U_3O_8	*Cd* Shield		Number of Counts per 30 Min.	Difference of Counts per 30 Min.	
		Outer	*Inner*			
C—i	present	with	no	2166		
D—i	„	with	with	2117	$(C-D)_i$	49
C—ii	„	with	no	2261		
D—ii	„	with	with	2155	$(C-D)_{ii}$	106
A	no	no	no	5633		
A'	„	semi-circular cylinder	no	5247	*A'—B*	2801
B	„	with	no	2446	*A—B*	3187
E	present	no	no	5478		
F	„	no	with	2436	*E—F*	3042

I) of the frequency of pulses per 30 minutes was found, when uranium oxide was irradiated by the thermal neutrons in the absence of the inner shield, and another count D-*ii* ($=2155$, in Table I) of pulses, when uranium oxide was screened from the thermal neutrons in the presence of the inner shield. The difference which must be attributed to the fast neutron emission, appears now to increase to the considerably large amount of *106* pulses. Here it is evident that there is a net increase in the number of the secondary fast neutrons as was presumed.

In the method of detection here adopted, there will be no occasion to take the reaction $(n, 2n)$ into account as Fermi and others[4] did, because, if there is any, it is sure to be canceled out by taking the difference of the numbers of counts for each pair of the alternating observations.

Now, in order to know the number of the primary thermal neutrons, to which uranium was exposed from the side of the semi-circular cylindrical surface of the paraffin block, the alternating change of the counts—with and without the outer cadmium cylinder—was measured for the empty uranium vessel, when the thermal neutrons were allowed to project towards the outer cadmium cylinder by removal of the inner shield during the measurements.

A count A ($=5633$, in Table I) taken in the absence of the outer cylinder, gave the frequency of pulses caused by the total primary—thermal and nonthermal—neutrons radiated from the whole cylindrical side of the paraffin block. Another count A' ($=5247$, in Table I) was taken with a cadmium shield of semi-circular cylindrical form covering only the back half side of the vessel, so that the measure obtained contains the number of the primary thermal neutrons emitted only from the fore half side of the cylindrical surface of the paraffin block. Further, a count B ($=2446$, in Table I) was a measure of pulses in the case when the detector had been shut off from the total primary thermal neutrons by setting the outer whole circular cylindrical shield.

Both the differences, $(A'-B)=2801$ and $(A-A')=386$ of the counts then will enable us to separate the group of the primary thermal neutrons emitted from the fore half side and that of the thermal neutrons from the back half side of the cylindrical surface of the paraffin block respectively.

In this way, we found i) 1.8 percent increase of the secondary

neutrons upon this primary in the first case, and ii) 3.8 percent increase in the second case. These increases must be regarded as caused only by the secondary emission of *nonthermal* neutrons due to the absorption of *thermal* neutrons alone.

It seems now reasonable to consider that the remarkable difference of these increases of counts observed between the first and second cases may be associated with probably *high* energies of the secondary neutrons. The secondary neutrons emitted in the process concerned are considered in the *second* case to be more strongly slowed down by passing through a part of the medium of the paraffin cylindrical block 20 cm in diameter; otherwise it might happen in the first case to realize a higher yield in the net increase of the secondary neutrons on account of its apparently superior geometrical conditions.

Energy of Secondary Neutrons liberated from Uranium

In order to estimate the energies of the secondary fast neutrons, a set of paraffin sheets (30 × 26 cm, 37 mm thick, each) was interposed between uranium oxide and the ionization chamber as seen in Fig. III, and the alternating change of the counts—with and without the inner cadmium shield—was observed as in the previous cases, for every different thickness of the increased paraffin layers.

Under the same geometrical arrangement as in the first case (i), layers of the paraffin sheet 37, 74 and 111 mm thick respectively were interposed between uranium oxide and the circular lead block, so that

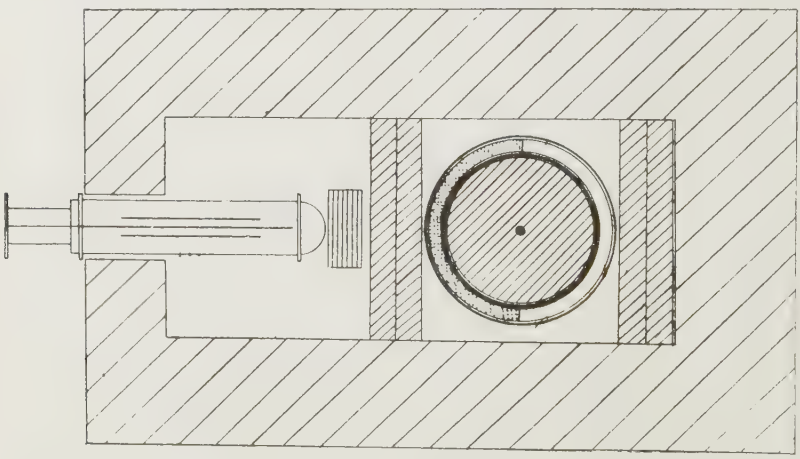

Fig. III. The paraffin sheets are interposed between the uranium oxide and the ionization chamber.

the nearest outside of the uranium part stood across the paraffin layers at a distance of 101, 138 and 175 mm respectively from the head of the ionization chamber.

Under the same geometrical arrangement as in the second case (ii), we have measured in two sets of experiments, in which the counts were recorded either in the presence or absence of only a single layer of the interposed paraffin sheet (37 mm thick), the nearest side of the uranium vessel being kept at a distance 101 mm from the head of the ionization chamber in its axial direction. The results obtained in the first and second cases are summarized in Table II and III respectively.

Table II

	Cd Shield		Thickness of Paraffin Layer	Number of Counts per 30 Min.	Difference of Counts: *C*—*D* per 30 Min.
	Outer	*Inner*			
C_1—*i*	with	no	0	2166	
D_1—*i*	with	with	„	2117	49
C_2—*i*	with	no	37 mm	2207	
D_2—*i*	with	with	„	2145	62
C_3—*i*	with	no	74 mm	1408	
D_3—*i*	with	with	„	1350	58
C_4—*i*	with	no	111 mm	786	
D_4—*i*	with	with	„	791	−5

Table III

	Cd Shield		Thickness of Paraffin Layer	Number of Counts per 30 Min.	Difference of Counts: *C*—*D* per 30 Min.
	Outer	*Inner*			
C_1—*ii*	with	no	0	2261	
D_1—*ii*	with	with	„	2155	106
C_2—*ii*	with	no	0 (*Uranium part is moved back 37 mm*)	2121	
D_2—*ii*	with	with	„	2019	102
C_3—*ii*	with	no	37 mm	2304	
D_3—*ii*	with	with	„	2256	48

The results in the first case (i) show that the increased numbers of the secondary fast neutrons appear notably larger after passing through the paraffin layers 37 and 74 mm thick, while the number falls rapidly

in the case of the last thickness of 111 mm. Thus it seems probable that the number of the secondary neutrons first increases with the increased paraffin sheets and then after passing through a maximum it decreases.

The results in the second case (ii) indicate that the number of the secondary neutrons is reduced already to about one half through a paraffin sheet 37 mm thick, while the removal of this paraffin sheet exerts no appreciable influence on the predominant increase of the original count, $(C\text{-}ii - D\text{-}ii) = 106$. (under the circumstance of the uranium vessel being separated by 101 mm, instead of 64 mm, from the neutron detector).

These facts can only be regarded as clear evidence of high energies of the fast neutrons concerned. Because of the lack of precision, however, it is inadequate from these results only to draw directly any numerical value for this problem.

In view of the results obtained, however, we have some reason[11] to conclude that the secondary fast neutrons emitted from uranium exposed to thermal neutrons possess a kinetic energy of *about 10 Mev.* in order of magnitude. This leads to the recognition of a wide departure from our early view that the order of a large amount of energy, such as about 100 Mev., might be expected to be available as kinetic energy for the secondary neutrons.

In view of the present evidence of the neutron emission with such an order of the amount of energy involved, we might justly say that the neutron release takes place *immediately* after the main division of the compound nucleus rather than simultaneously with the explosion.

Delayed Emission of Neutrons from Uranium

Roberts, Meyer and Wang[12] and Booth, Dunning and Slack[13] have reported neutron emission *delayed* for about 12 sec. and 45 sec. periods, in the case when uranium is irradiated.

We also looked for such delayed fast neutrons in the *present arrangement*, but observed no positive phenomena which might be due to the delayed emission[14] after the quick removal of the source from

11. K. Kimura, Memoirs of the College of Science, Kyoto Imperial University, A, **22**, 241 (1939).

12. R. B. Roberts, R. C. Meyer and P. Wang, Phys. Rev., **55**, 510 (1939). R. B. Roberts, L. R. Hafstad, R. C. Meyer and P. Wang, Phys. Rev., **55**, 664 (1939).

13. E. T. Booth, J. R. Dunning and F. G. Slack, Phys. Rev., **55**, 876 (1939).

14. Further detailed evidence regarding the delayed neutrons is to be reported by other investigators in this laboratory.

the paraffin block. So it is to be considered at least that the number of the delayed emission may be much less[4,6] than the number of those secondary neutrons which have been projected into the detector during the direct irradiation of the primary neutrons, and if there is any at all, it seems actually too small to be observable in the present neutron measurements.

Possibility of Nuclear Chain Reactions

In the preceding experiments it was directly found that there is an emission of fast neutrons from uranium under the irradiation by thermal neutrons alone.

In order to detect whether or not the number of the fast neutrons so released actually exceeds the total number of the thermal neutrons absorbed in uranium oxide by all processes whatever, under the action of thermal neutrons, it is necessary to know a measure for the decrease due to the total absorption of thermal neutrons by uranium oxide layer in the same units of the present experiment. The experiments were carried out in the following way.

As was formerly done, the effects of the presence (F in Table I) and absence (E in Table I) of the inner cadmium shield were measured, other conditions being the same as in the first experiment except that the outer cadmium shield was removed from the outside of the vessel during the experiments. The difference between these two counts will give a total measure of the transmission of the thermal neutrons through uranium oxide, including even the neutrons due to the emission of the fast neutrons caused by the fission process. The fraction of the actual transmission of the primary thermal neutrons, therefore, would correspond to a number of count, $(E-F)-(C-D)-(A-A')$, which has been obtained by subtracting the fraction of the secondary emission of fast neutrons from the above total transmission and further by taking into account a fraction of the primary neutrons which, in the present arrangement, would go round back even to the neutron detector as the opposite component emitted from the back half side of the circular cylindrical surface of the paraffin block.

The difference between the number, $(A'-B)$, of the thermal neutrons emitted by the source and the number, $(E-F)-(C-D)-(A-A')$, of the neutrons transmitted through uranium oxide is due to the absorption of the thermal neutrons by uranium oxide. The total absorption cross section σ_{tot} of uranium oxide due to fission and other

possible processes in the present experimental arrangement was calculated from these results to be

$$\sigma_{tot} = 9.6 \times 10^{-24} \text{ cm}^2,$$

by use of the relation of the exponential absorption.

In this way, it was found that 7 percent of the primary thermal neutrons emitted by the source is absorbed by uranium oxide. Accordingly, if all these thermal neutrons absorbed were assumed to participate in the fission process alone, the observed increase of the count, 3.8 percent, would correspond apparently to no more than an average emission of about 0.6 fast neutrons for each thermal neutron absorbed.

This result means that the number of the secondary fast neutrons produced does not exceed the total number of the thermal neutrons absorbed within uranium oxide by all possible sorts of capture processes, and that the production of a chain of nuclear reactions would not be expected in the *ordinary* uranium oxide exposed to thermal neutrons alone.

Average Number of Secondary Neutrons produced per Fission of Uranium

In order to estimate the *average number* ν of the secondary fast neutrons liberated per fission of uranium induced by thermal neutron, let σ_f and a_f be the cross section and the absorption coefficient of the thermal neutrons respectively for this fission process alone, n the number of the nuclei per unit volume and r the path length of neutrons travelling through uranium oxide. Then, $\nu a_f r$, or $\nu \sigma_f n r$ should correspond to the ratio of the number, $(C-D)$, of the observed net gain of the secondary neutrons from uranium oxide to that, $(A'-B)$, of the thermal neutrons emitted by the source, *i.e.*, to $\left(\frac{k'}{k}\right)\cdot\frac{C-D}{A'-B}$, where k and k' are the average efficiency factors of the detector for *slow* neutrons and for *fast* neutrons respectively, which have been projected from the outer cadmium surface of the uranium container.

It is here assumed that the contributions of all fast and slow neutrons of different energies to the detecting efficiency of the neutron detector in the paraffin chamber are of the same order of magnitude. But it may be generally difficult to realize such a condition in the practical arrangement used, and the above correlation will hold only approximately as it might give either too low or too high a value, according as $\frac{k'}{k} \lesseqgtr 1$, for the number of ν.

Since, however, in the geometry of the previous case (ii) at least, it is found to be possible to obtain with good approximation a condition similar to that required above, putting $k \fallingdotseq k'$, we have

$$\nu = \frac{1}{\sigma} \cdot \frac{C-D}{A'-B}, \qquad \text{where} \quad \sigma = nr\sigma_f.$$

If we assume the observed value of σ_f for thermal neutrons—the *average* value over the actual mixture of isotopes for the oxide,—given by Anderson, Booth, Dunning, Fermi, Glasoe and Slack,[15] we have $\sigma_f = 2 \times 10^{-24}$ cm^2. Since $\frac{C-D}{A'-B} = 0.038$, we find :

$$\nu = 2.5.$$

This would be, however, an average number of neutrons released per fission if the intensities of both the incident thermal neutrons and the fission neutrons produced were not reduced by the absorption along the travelling path length of the uranium oxide layer before they escaped from the whole mass.

By taking into account the effects of this reduction in the number of the neutrons within uranium oxide, this average number of ν must be increased by a certain fraction. Assuming the exponential law for these neutrons in such absorber, the magnitude of this correction factor is to be given by the following expression :

$$\frac{(\sigma_{tot} - \sigma_{fast})nr}{e^{-\sigma_{fast}nr}\left\{1 - e^{(\sigma_{fast} - \sigma_{tot})nr}\right\}},$$

where σ_{tot} is the total absorption cross section for all possible capture processes of thermal neutrons, σ_{fast} the corresponding quantity for the capture of the fast fission neutrons produced. Putting the numerical values of the quantities necessary[16] in the above expression, the value of the factor is estimated to be 1.05. Accordingly we find

$$\nu = 2.6.$$

Conclusion

Thus, by the present investigation it was made possible to decide

15. H. L. Anderson, E. T. Booth, J. R. Dunning, E. Fermi, G. N. Glasoe and F.G. Slack, Phys. Rev., **55**, 511 (1939).

16. For the estimation of the correction factor, the value of σ_{fast} is assumed to be comparable to that of the next heavy elements ($\sigma_{fast} \simeq 2.5 \times 10^{-24}$ cm^2) for fast neutrons. ($E \geq 7$ Mev.)—D. C. Grahame and G. T. Seaborg, *Phys. Rev.*, **53**, 797 (1938).

conclusively the remarkable *net increase* of the secondary fast neutrons caused only by the thermal neutrons absorbed, and also to estimate that the *mean number of* ν is 2.6, as deduced above.

Considering an order of the amount of energy—about 10 Mev.—involved in the fission neutrons, it is probable that the neutron release takes place *immediately* after the main division of the compound nucleus instead of simultaneously with the explosion.

The author is deeply indebted to Prof. B. Arakatsu for his helpful suggestions and invaluable advice so generously given during the course of this work, and to other members of the nuclear research laboratory for their active interest. His thanks are due to the Ensuiko-Sugar Company for the loan of the neutron source and to the Nippon Gakujutsu Shinkokwai for their financial support. He also wishes to express his deepest gratitude to Prof. S. Horiba for the continual interest he has taken throughout the work.

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October, 1939.

Note added in proof:—Since this manuscript was submitted for publication, a report by Mr. W. H. Zinn and L. Szilard, *Phys. Rev.*, **56**, 619 (1939), has appeared, in which the same problem was studied by recording photographically helium and hydrogen recoils due to the fission neutrons. Their experiment differs from the present work in the method of neutron detection adopted. Their value obtained for the number of neutrons emitted per fission, however, is in agreement with the author's result.

Vol. XXIII, No. 1, Published, January 31, 1940

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昭和十五年一月二十五日印刷
昭和十五年一月三十一日發行

編纂兼發行者　京都帝國大學理學部

印刷者　須磨勘兵衛
京都市下京區北小路通新町西

印刷所　內外出版印刷株式會社
京都市下京區西洞院通七條南

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